

AMENDMENTS TO THE CLAIMS

1. (Currently Amended) A catalyst system capable of catalysing the carbonylation of an olefinally unsaturated compound, which catalyst system is obtainable by combining:

(a) ~~(e)~~ a metal of Group VIB or Group VIII B or a compound thereof: and

(b) ~~(d)~~ a bidentate phosphine of general formula (I)

~~(I)~~



wherein:

Ar is a bridging group comprising an optionally substituted aryl moiety to which the phosphorus atoms are linked on available adjacent carbon atoms;

A and B each independently represent lower alkylene;

K, D, E and Z are substituents of the aryl moiety (Ar) and each independently represent hydrogen, lower alkyl, aryl, Het, halo, cyano, nitro, OR^{10} , OC(O)R^{11} , C(O)R^{12} , C(O)OR^{13} , $\text{NR}^{14}\text{R}^{15}$, $\text{C(O)NR}^{16}\text{R}^{17}$, $\text{C(S)R}^{16}\text{R}^{17}$, SR^{18} , C(O)SR^{18} , or $-\text{J-Q}^3(\text{Ad})_w(\text{CR}^7(\text{R}^8)(\text{R}^9))_x$ where J represents lower alkylene; or two adjacent groups selected from K, Z, D and E together with the carbon atoms of the aryl ring to which they are attached form a further phenyl ring, which is optionally substituted by one or more substituents selected from hydrogen, lower alkyl, halo,

cyano, nitro, OR^{10} , OC(O)R^{11} , C(O)R^{12} , C(O)OR^{13} , $\text{NR}^{14}\text{R}^{15}$, $\text{C(O)NR}^{16}\text{R}^{17}$, $\text{C(S)R}^{16}\text{R}^{17}$, SR^{18} or C(O)SR^{18} ;

R^1 to R^6 each independently represent lower alkyl, aryl, or Het;

Ad each independently represent an optionally substituted adamantyl radical bonded to the phosphorous atom via any one of its tertiary carbon atoms, the said optional substitution being by one or more substituents selected from hydrogen, lower alkyl, halo, cyano, nitro, OR^{10} , OC(O)R^{11} , C(O)R^{12} , C(O)OR^{13} , $\text{NR}^{14}\text{R}^{15}$, $\text{C(O)NR}^{16}\text{R}^{17}$, $\text{C(S)R}^{16}\text{R}^{17}$, SR^{18} or C(O)SR^{18} ;

R^{10} to R^{18} each independently represent hydrogen, lower alkyl, aryl or Het;

$\text{S} \ \& \ \text{U} = 0, 1 \text{ or } 2$ provided that $\text{S} + \text{U} \geq 1$;

$\text{T} \ \& \ \text{V} = 0, 1 \text{ or } 2$ provided that $\text{T} + \text{V} \leq 3$;

$\text{W} \ \& \ \text{X} = 0, 1 \text{ or } 2$;

Q^1 , Q^2 and Q^3 (when present) each independently represent phosphorous, arsenic or antimony and in the latter two cases references to phosphine or phosphorous above should be varied accordingly.

2. (Original) A catalyst system according to claim 1 wherein, the Group VIII B metal is palladium.

3. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, wherein R¹ to R⁹ each independently represent lower alkyl, aralkyl or aryl.

4. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, wherein R¹ to R⁹ each independently represent C₁ to C₆ alkyl, C₁-C₆ alkyl phenyl (wherein the phenyl group is optionally substituted as defined herein) or phenyl (wherein the phenyl group is optionally substituted as defined herein).

5. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, wherein each Q¹, Q² and Q³ (when present) are the same.

6. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, each Q¹, Q² and Q³ (when present) represents phosphorous.

7. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, wherein A, B and J (when present) each independently represent C₁ to C₆ alkylene which is optionally substituted as defined herein, for example with lower alkyl groups.

8. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, wherein when K, D, E or Z does not represent -J-Q³(Ad)_w(CR⁷(R⁸)(R⁹))_x, K, D, E or Z represents hydrogen, lower alkyl, phenyl or lower alkylphenyl.

9. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, wherein when K, D, E and Z together with the carbon atoms of the aryl ring to which they are attached do not form a phenyl ring, K, D, E and Z each independently represent hydrogen, lower alkyl, phenyl or lower alkylphenyl.

10. (Currently Amended) A catalyst system according to claim 1 ~~any of claims 1-8~~, wherein when two of K, D, E and Z together with the carbon atoms of the aryl ring to which they are attached form a phenyl ring, then the phenyl ring is optionally substituted with one or more substituents selected from aryl, lower alkyl (which alkyl group may itself be optionally substituted or terminated as defined below), Het, halo, cyano, nitro, OR^{10} , $OC(O)R^{11}$, $C(O)R^{12}$, $C(O)OR^{13}$, $NR^{14}R^{15}$, $C(O)NR^{16}R^{17}$, SR^{18} , $C(O)SR^{18}$ or $C(S)NR^{16}R^{17}$ wherein R^{10} to R^{18} each independently represent hydrogen or lower alkyl (which alkyl group may itself be optionally substituted or terminated as defined herein).

11. (Currently Amended) A catalyst system according to claim 1 ~~any preceding claim~~, wherein $S \geq 1$ and $u \geq 1$.

12. (Currently Amended) A process for the carbonylation of an ethylenically unsaturated compound comprising contacting an ethylenically unsaturated compound with carbon monoxide and a hydroxyl group containing compound in the presence of a catalyst system in accordance with Claim 1 ~~any of claims 1-11~~.

13. (Original) A process according to claim 12, wherein the hydroxyl group containing compound includes water or an organic molecule having a hydroxyl functional group.

14. (Original) A process according to claim 13, wherein the organic molecule having a hydroxyl functional group may be branched or linear, and comprises an alkanol, particularly a C₁-C₃₀ alkanol, including aryl alkanols, which may be optionally substituted with one or more substituents selected from lower alkyl, aryl, Het, halo, cyano, nitro, OR¹⁰, OC(O)R¹¹, C(O)R¹², C(O)OR¹³, NR¹⁴R¹⁵, C(O)NR¹⁶R¹⁷, C(S)R¹⁶R¹⁷, SR¹⁸ or C(O)SR¹⁸ as defined herein.

15. (Currently Amended) A process according to claim 12 ~~any of claims 12-14~~, wherein the carbonylation of an ethylenically unsaturated compound is performed in one or more aprotic solvents.

16. (Currently Amended) A process according to claim 12 ~~any of claims 12-14~~, wherein the reaction is carried out in the absence of any external added aprotic solvent ie. an aprotic solvent not generated by the reaction itself.

17. (Currently Amended) A process according to claim 12 ~~any of claims 12-16~~, wherein the anion may be derived from or introduced as one or more of an acid having a pKa measured in aqueous solution at 18°C of less than 4.

18. (Currently Amended) A process according to claim 1 ~~any of claims 12-17~~, wherein suitable ethylenically unsaturated compounds include ethene, propene, hexene, vinyl compounds

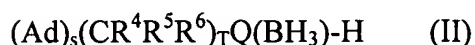
such as vinyl acetates, heptene, octene, nonene, decene, undecene, dodecene, etc up to C₃₀ which may be linear or branched, cyclic or uncyclic or part cyclic and in which the double bond may take any suitable position in the carbon chain and which includes all stereoisomers thereof.

19. (Currently Amended) A catalyst system according to claim 1 ~~any of claims 1-11~~, wherein the catalyst system includes in a liquid reaction medium a polymeric dispersant dissolved in a liquid carrier, said polymeric dispersant being capable of stabilising a colloidal suspension of particles of the group VI or VIIIB metal or metal compound of the catalyst system within the liquid carrier.

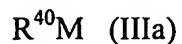
20. (Currently Amended) A catalyst system according to claim 1 ~~any of claims 1-11 or 19~~, wherein each of the groups R¹ to R³, R⁴ to R⁶ and R⁷ to R⁹ together independently may form cyclic structures such as 1-norbornyl or 1-norbornadienyl.

21. (Currently Amended) A method of preparation of an intermediate for a bidentate phosphine of general formula (I) in accordance with claim 1 ~~any of claims 1-11~~, which comprises the steps of:

reacting the borane protected moiety of formula (II)



wherein Ad, R⁴, R⁵, R⁶, Q, S and T are as described previously and H is a hydrogen atom;
with a compound of formula (IIIa)



Or IIIb



wherein R^{40} is a branched $C_1 - C_8$ alkyl group joined to the metal M at a secondary or tertiary carbon and M represents a group IA alkali metal and

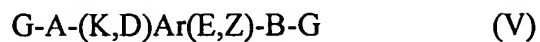
wherein $R^{41}-M$ is generated in situ by an exchange reaction between $R^{41}-Li$ and KOR^{42} or $NaOR^{42}$, wherein R^{41} and R^{42} are independently $C_1 - C_8$ alkyl, aryl or aralkyl groups which may be linear or branched to produce



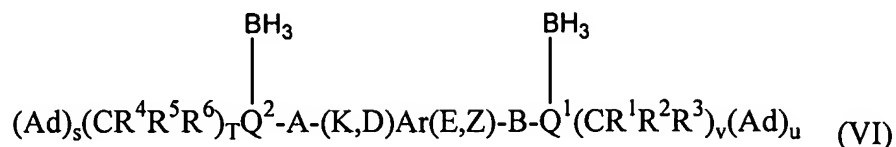
22. (Original) A method according to claim 21, wherein R^{40} is sec-butyl or tert-butyl.

23. (Currently Amended) A method according to claim 21 ~~or 22~~, wherein M is lithium.

24. (Currently Amended) A method according to claim 21 ~~any of claims 21 to 23~~, wherein the intermediate of formula IV is reacted with the bridging moiety V



wherein A, K, D, Ar, E, Z and B are as already defined and G is a halogen radical, preferably chlorine; to produce



wherein Q^1 may be the same or different than Q^2 .

25. (Original) A method according to claim 24, wherein the method includes the further step of borane deprotection.

26. (Original) A method according to claim 25, wherein the borane deprotection is effected with tetrafluoroboric acid dimethyl ether complex.

27. (Original) A method according to claim 21 ~~any of claims 21 to 23~~, wherein the reaction takes place at less than 80°C, preferably less than 50°C.

28. (Original) A method according to claim 24, wherein the reaction takes place at less than 80°C, preferably less than 50°C.

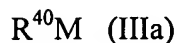
29. (Currently Amended) A method of preparation of an intermediate for a bidentate phosphine of general formula (I) in accordance with claim 1 ~~any of claims 1-11~~, which comprises the steps of:

reacting the borane protected moiety of formula (II)



wherein Ad, R^4 , R^5 , R^6 , Q, S and T are as described previously and H is a hydrogen atom;

with a compound of formula (IIIa)



Or IIIb



wherein R^{40} is a $\text{C}_1 - \text{C}_8$ alkyl, aryl or aralkyl group, M represents a group IA alkali metal and $\text{R}^{40}\text{-H}$ has a pK_a greater than n-butane and

wherein R^{41} -M is generated in situ by an exchange reaction between R^{41} -Li and KOR^{42} or $NaOR^{42}$, wherein R^{41} and R^{42} are independently a $C_1 - C_8$ alkyl, aryl or aralkyl group which may be linear or branched.

30. (Original) A method according to claim 29, wherein R^{40} , R^{41} and R^{42} are independently selected from a C^1 - C^8 alkyl or aralkyl radical.